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Spin waves in terbium. I. Two-ion magnetic anisotropy

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The energies of spin waves propagating in the c direction of Tb have been studied by inelastic neutron scattering, as a function of a magnetic field applied along the easy and hard directions in the basal plane, and as a function of temperature. From a general spin Hamiltonian, consistent with the symmetry, we deduce the dispersion relation for the spin waves in a basal-plane ferromagnet. This phenomenological spin-wave theory accounts for the observed behavior of the magnon energies in Tb. The two $\vec{q}$-dependent Bogoliubov components of the magnon energies are derived from the experimental results, which are corrected for the effect of the direct coupling between the magnons and the phonons, and for the field dependence of the relative magnetization at finite temperatures. A large $\vec{q}$-dependent difference between the two energy components is observed, showing that the anisotropy of the two-ion coupling between the magnetic moments in Tb is substantial. The $\vec{q}$-dependent anisotropy deduced at 4.2 K is of the same order of magnitude as the isotropic part, and depends strongly on the orientation of the moments in the basal plane. The rapid decrease of both the axial- and the basal-plane anisotropy with increasing temperatures implies that the two-ion coupling is effectively isotropic above ~150 K. We present arguments for concluding that, among the mechanisms which may introduce anisotropic two-ion couplings in the rare-earth metals, the modification of the indirect exchange interaction by the spin-orbit coupling of the conduction electrons is of greatest importance.

I. INTRODUCTION

The present series of articles with the common title "Spin waves in terbium" and with the subtitles "Two-ion magnetic anisotropy," "Magnon-phonon interaction," and "Magnetic anisotropy at zero wave vector" (to be referred to as I, II, and III, respectively), constitute together a comprehensive study of the magnetic excitations in the ferromagnetic phase of Tb.

The magnetic behavior of the rare-earth metals is basically understood by making a sharp distinction between the localized $4f$ electrons carrying essentially all the magnetic moment and the conduction electrons. The strong coupling between the spins and the orbital momentum of the electrons in the unfilled $4f$ shell makes it possible to treat the total spin $\vec{S}$, the orbital momentum $\vec{L}$, and the angular momentum $\vec{J}=\vec{L}+\vec{S}$ of the $4f$ electrons as constants of motion. The electric field from the surroundings acting on the $4f$ electrons of the trivalent rare-earth ions in the metal is screened by the filled $5s$ and $5p$ shells and is weak in comparison with the spin-orbit coupling. The ground-state J multiplet of the trivalent ions, which in the case of Tb is determined by $S=L=3$ and $J=6$ (as may be obtained by the application of Hund's rules) is then responsible for the magnetic properties of the rare-earth metals.

Cooperative phenomena occur in the rare-earth metals because the moments on different sites are coupled together quite strongly through the conduction electrons.[1] In the heavy-rare-earth series (Gd–Tm) the two-ion coupling dominates the crystal field, and the mixing of different M_J states may be treated as a perturbation. In general, the magnetic excitations in the ordered phases of the heavy-rare-earth metals (in Er and Tm only in the low-temperature phases) are considered to be well-defined spin waves for temperatures well below the transition temperatures. The mixing of different M_J states will then appear as magnetic anisotropy, or as a tendency of the moments to be oriented along a preferred direction, and it will give rise to zero-point deviations from the fully aligned ground state assumed in a spin-wave approach. The presence of magnetic anisotropy will manifest itself in the energy of the magnons and most distinctly as an energy gap at the wave vector $\vec{Q}$ of a periodic magnetic structure ($\vec{Q}$ is zero for a ferromagnet). Finally, the M_J mixing will influence the magnon-magnon interaction and thus contributes to the renormalization of the energies and to the lifetime of the magnons.

A nonvanishing orbital momentum induces a coupling between spin space and real space, and the large values of L which occur in the heavy rare earths, with the exception of Gd^{+3} (^8S ground state), are responsible for the strongly anisotropic behavior observed in these metals. In the ferromagnetic phase of Gd the static anisotropy, the energy gap at $q=0$, and the interaction between the spin system and the lattice are at least an order of magnitude smaller than in the other heavy-rare-earth metals.

In the present paper and the two subsequent ones we study in detail different effects of magnetic anisotropy on the spin waves in ferromagnetic Tb. The crystal structure of Tb (and of the other heavy rare earths) is hexagonal close packed, and Tb is ferromagnetically ordered below $T_C = 216$ K, with the moments lying in the basal plane along a b axis. The moments can be oriented along a hard a direction by the application of an external field (~30 kOe at 4.2 K) in this direction.

Having reviewed the kinds of spin interactions which may occur in the rare-earth metals, we introduce a general spin Hamiltonian which includes all imaginable single-ion and two-ion couplings as well as magnetoelastic couplings. By a fairly simple approach we derive a general dispersion relation valid for spin waves propagating in the c direction of a basal-plane ferromagnet. In directions in the Brillouin zone other than the highly symmetric c direction, additional contributions may be present. The effect of an external field on the dispersion relation is considered, and the renormalization of the energies at finite temperatures is included in an effective fashion.

We have studied experimentally the spin-wave dispersion relation in the c direction of Tb by the technique of inelastic neutron scattering. The presence of $\vec{q}$-dependent two-ion anisotropy is reflected in the field dependence of the magnon energies. By measuring the energies as a function of temperature and of a magnetic field applied along both the easy and hard directions, we found that anisotropic two-ion couplings are important in a description of the magnetic properties of Tb. The $\vec{q}$-dependent anisotropy which we deduce depends strongly on the direction of the magnetization in the basal plane and decreases quite rapidly when the temperature is increased. The experimental results were corrected for the perturbation due to magnon-phonon interaction and for the influence of the field dependence of the relative magnetization at finite temperatures.

The $\vec{q}$-dependent anisotropy which is obtained after having isolated the effects of the direct couplings between magnons and phonons is related more closely to pure spin interactions. In the next paper (II) the selection rules for the linear couplings between magnons and phonons propagating in the c direction of a basal-plane ferromagnet are determined. The spin waves propagating in the c direction in Tb are coupled to both the acoustic and optical branches of the transverse-phonon spectrum. The experimental studies of the acoustic-optical coupling indicate that this interaction, which is the largest one observed in Tb, violates these selection rules. The main properties of the spin (and phonon) system of Tb seem to be well understood, and we consider this problem to be one which is connected to the coupling mechanism itself. We propose in II that the acoustic-optical interaction violates the selection rules for a simple ferromagnet because of the deviation between the direction of magnetization and that of the conduction-electron polarization, which is proportional to the spin-orbit coupling.

Finally, in the last paper (III), we concentrate on the behavior of the energy gap at zero wave vector. From the measurements of the field dependence of the energy gap the dynamic anisotropy parameters are deduced as a function of temperature. There is convincing evidence for large two-ion contributions to the magnetic anisotropy at zero wave vector. The dynamic parameters deduced are compared with the corresponding static values measured by other experiments.

II. SPIN INTERACTIONS

The most important two-ion coupling between the ionic magnetic moments of a rare-earth metal is generally assumed to be due to overlap of the wave functions of the conduction electrons and of the localized $4f$ electrons. The exchange between the itinerant and the localized electrons then leads to a coupling of the spins on different sites mediated by the propagation of the itinerant electrons. Assuming the spin-orbit splitting to be much larger than the exchange energy and the response function (the susceptibility) of the conduction electrons to be isotropic in space, this coupling takes the form of an isotropic Heisenberg interaction, when orbital effects are neglected,

$$\mathcal{H}_H = -\tfrac{1}{2}\sum_{i \neq j} \mathcal{J}(\vec{R}_i - \vec{R}_j)\vec{J}_i \cdot \vec{J}_j \,, \tag{1}$$

where $\vec{J}_i$ is the total angular moment on site $\vec{R}_i$. This simple indirect exchange mechanism, the so-called Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction,[1] seems to give a generally satisfactory description of the magnetic ordering in the rare-earth metals. The strong coupling between the conduction electrons and the local moments is clearly demonstrated by the effect of magnetic ordering on the electronic properties.[2] The transition temperatures are found to vary roughly linearily with the Néel-de Gennes factor[3]

$$G = (g-1)^2 J(J+1) \tag{2}$$

across the heavy rare earths and a wide range of alloys[4] (g is Landé's factor), as would be expected for a pure spin-spin interaction. The two-ion coupling expresses itself most directly in the dispersion of the magnetic excitations of the system, which can be studied by inelastic neutron scattering. However, it is in general not possible to deduce the actual form of the two-ion spin Hamiltonian from the dispersion relation only.

Various orbital effects contribute to the spin Hamiltonian and the two-ion coupling may therefore be anisotropic. In this case it is more convenient to express the Hamiltonian in terms of Racah operators,[5,6] $\tilde{O}_{l,m}(J_i)$, which are tensor operators corresponding to the spherical harmonics and characterized by simple transformation properties. In this notation $\vec{J}_i \cdot \vec{J}_j$ in (1) is replaced by

$$\vec{J}_i \cdot \vec{J}_j = \sum_{m=-1,0,1} (-1)^m \tilde{O}_{1,m}(J_i)\tilde{O}_{1,-m}(J_j). \tag{3}$$

With the purpose of deriving a general expression for the spin waves propagating in the c direction of Tb we shall begin by considering a general spin interaction term

$$\mathcal{K}^{\mu mm'}_{\lambda ll'}(\vec{R}_i - \vec{R}_j)\langle \tilde{O}_{\lambda,\mu}\rangle \tilde{O}_{l,m}(J_i)\tilde{O}_{l',m'}(J_j), \tag{4}$$

where we have introduced the effect of the surroundings as a mean field. The effects of a possible spatial dependence of the expectation value of the Racah operator, $\tilde{O}_{\lambda,\mu}$, are neglected ($\langle\ \rangle$ then includes an averaging in space of the expectation value).

In the rare-earth metals, several kinds of mechanisms may introduce anisotropic two-ion couplings, all of which can be written in the form given by Eq. (4). The exchange interaction may give rise to other terms than the Heisenberg interaction as listed below, (i)–(v). In addition to the exchange interaction other types of spin interactions are present in the rare-earth metals, (vi)–(ix), among which the most important are those with their origin in the Coulomb interaction between the ions:

(i) In the case where the ions possess an orbital moment the s-f exchange interaction is influenced by changes in the orbital moment of the conduction electrons (with respect to the ions) during the scattering process. This effect has been treated by Kaplan and Lyons[7] and others[8,9] using a number of simplifying assumptions. Kaplan and Lyons obtained, as a modification of the Heisenberg Hamiltonian, a pseudodipolar interaction with a magnitude of the order of 10% of the isotropic interaction.

(ii) The spin-orbit coupling modifies the wave functions of the conduction electrons and thus the s-f exchange matrix element. This mixing of the wave functions, as considered in some detail by Levy,[10] may introduce anisotropic couplings, which differ from those due to the orbit-orbit coupling, (i), by the possible existence of nonvanishing anisotropy in the case of S-state ions (Gd^{+3}) and of antisymmetric couplings[11,12] which change sign when i and j are interchanged in (4). The order of magnitude relative to the exchange energy of the spin-orbit terms is roughly determined by the ratio of half the spin-orbit splitting to the d-band width, $\langle \lambda_{\text{s-o}} \rangle$, as estimated by Liu[13] in another context to be equal to 0.03.

(iii) As a simple consequence of the dependence of the exchange interaction on the distance between the ions $\vec{R}_i - \vec{R}_j$, the spin system is coupled to the phonons. This coupling does not give rise to anisotropy itself, but it may strengthen existing anisotropy [see also (viii)]. In contrast to (i) and (ii) this interaction introduces magnetostrictive terms for which λ in (4) is nonzero.

(iv) When the crystal-field splitting is small compared with the exchange energy, as in the heavy-rare-earth metals, the s-f exchange interaction perturbs the uniform spin-wave mode so as to give rise to a relative reduction of the energy of the order of 2.5% (in Tb).

(v) The s-f exchange interaction (1) is proportional to the susceptibility of the conduction electrons. Owing to the polarization and the spin-orbit coupling of the conduction electrons this susceptibility may be anisotropic in space and hence introduce terms where λ is nonzero.[14-16] The main effect is a difference between the susceptibility parallel and perpendicular to the magnetization,[16] which at zero wave vector is of the order $(\mu_B H_{\text{Ex}}/E_F)^2 \simeq 5\times10^{-5}$ in Tb for the polarization contributions, and $\langle\lambda_{\text{s-o}}\rangle^2 \simeq 10^{-3}$ for the spin-orbit contributions relative to the total static susceptibility (μ_B is the Bohr magneton, H_{Ex} is the exchange field, and E_F is the Fermi energy of the conduction electrons). Although these terms are small, they may have some detectable effects on the static anisotropy (III). At finite wave vector these contributions may be much larger; however, they are not directly observable because the dispersion of the magnetic excitations is independent of the parallel susceptibility.

The other very important source of anisotropy which occurs for the rare-earth ions is the dependence of the Coulomb interaction of two ions on the orbital states of the ions. This electrostatic interaction between the electronic charge clouds of the ions may be divided into two classes:

(vi) crystal-field terms ($l \neq 0$, l' and $\lambda = 0$) and electric multipole interactions (l and $l' \neq 0$, $\lambda = 0$) and (vii) the corresponding strain dependent terms in which λ is different from zero.

The Coulomb interaction may contribute only with terms in which λ, l, and l' are all even, if configuration mixing is neglected. The single-ion terms ($l' = 0$) are known to be quite large, giving rise to static anisotropy and to magnetostriction. The pure two-ion couplings ($l' \neq 0$), which have been considered by Wolf and Birgeneau,[17] are probably small; the most likely possibility is the quadropole-quadropole interaction,[18] where $l = l' = 2$. The direct Coulomb interaction between the localized $4f$ electrons and the conduction electrons[19] may have a pronounced effect on the behavior of the Coulomb terms. The strain dependent interactions, (vii) and (iii), give rise to an indirect two-ion coupling (corresponding to the indirect exchange interaction) by the excitation of virtual phonons:

(viii) Phonon-induced (electric) multipole interactions. This kind of two-ion coupling was first proposed by Sugihara[20] and has been treated more recently by Orbach and Tachiki[21] and others.[18,22] This effect is known to be very important in some rare-earth salts of the Jahn-Teller type.[22]

(ix) Besides the mechanisms quoted in (i)–(viii) we may have additional contributions to the spatially anisotropic two-ion couplings from (a) direct overlap[23] between the $4f$ wave functions, (b) indirect exchange via polarization of the $5s$ and $5p$ electrons,[24] (c) indirect interactions via the spin-orbit coupling of the conduction electrons[12] (independent of the exchange), and (d) interactions between the ground-state J multiplets and other J multiplets.[25] All these terms are presumably very small, but the sum of their contributions to the Hamiltonian may not be negligible.

Because of the great variation in possible contributions to the spin Hamiltonian, we will not *a priori* exclude any terms of the form given by (4). The actual form of the Hamiltonian is then determined by symmetry, see, e.g., Elliott and Thorpe[26] and the review by Wolf.[27] The angular dependence of a charge cloud may be expanded in spherical harmonics up to order $2l$, if l is the angular momentum of the electrons; so λ is ≤ 6 for $4f$ electrons ($l = 3$). The orbital dependence of the exchange interaction, (i)–(v), may introduce terms where l and l' in (4) are equal to 7:

$$\lambda \leq 6, \quad l \text{ and } l' \leq 7 \tag{5}$$

(two-ion magnetoelastic couplings may give rise to terms in which the effective λ is greater than 6). Time-reversal symmetry combined with the invariance of the system to Hermitian conjugation requires that

$$\lambda + l + l' \text{ be even,}$$

$$\mathcal{K}_{\lambda l l'}^{-\mu, -m, -m'}(\vec{R}_i - \vec{R}_j) = (-1)^{\mu+m+m'} \mathcal{K}_{\lambda l l'}^{\mu m m'}(\vec{R}_i - \vec{R}_j)^* . \tag{6}$$

The conditions (5) and (6) are general. The symmetry elements of the magnetic group impose further restrictions on the terms which may be present, and in Sec. III we shall consider an explicit example.

III. SPIN WAVES

The spin Hamiltonian for magnetic excitations propagating in the c direction of the heavy-rare-earth metals (hcp structure) can be reduced by the symmetry operations which leave the hexagonal layers unchanged. If the crystal is magnetically ordered, the use of these symmetry elements presupposes that the moments within a certain hexagonal layer are parallel and equal in magnitude (this condition is fulfilled in all the heavy rare-earth metals). The effective spin Hamiltonian obtained in this case can be written

$$\begin{aligned} \mathcal{H}_{MJJ} = \tfrac{1}{2} \sum_{i \neq j} \sum_{\lambda\mu} \sum_{lm} \sum_{l'm'} \frac{1}{S_\lambda S_l S_{l'}} & \times [K_{\lambda l l'}^{\mu m m'}(\vec{R}_i - \vec{R}_j) \langle \tilde{O}_{\lambda,\mu} \rangle \tilde{O}_{l,m}(J_i) \tilde{O}_{l',m'}(J_j) \\ & + (-1)^{\mu+m+m'} K_{\lambda l l'}^{\mu m m'}(\vec{R}_i - \vec{R}_j)^* \\ & \times \langle \tilde{O}_{\lambda,-\mu} \rangle \tilde{O}_{l,-m}(J_i) \tilde{O}_{l',-m'}(J_j)], \end{aligned} \tag{7}$$

where

$$\mu + m + m' = 6p, \quad p = 0, 1, 2, 3, \tag{8a}$$

when referring to a coordinate system with the x, y, and z axes along the a, b, and c directions, respectively. For simplicity we have neglected terms for which p is a half integer. These terms, which may occur because of the spin-orbit coupling of the conduction electrons [mechanism (ii)], introduce a coupling between acoustic and optical magnons proportional to $\cos 3n\phi$ ($n = 1, 3, \ldots,$ and ϕ is the angle the magnetization makes with the a axis). Although the acoustic-optical coupling may be finite at the Brillouin-zone boundary (A), the normal modes remain doubly degenerate at this point. The coupling may only be of importance close to A, and we shall henceforward neglect it. In that case we have further

$$\begin{aligned} K_{\lambda l l'}^{\mu m m'}(\vec{R}_i - \vec{R}_j)^* &= K_{\lambda l l'}^{\mu m m'}(\vec{R}_i - \vec{R}_j) \\ &= K_{\lambda l l'}^{\mu m m'}(\vec{R}_j - \vec{R}_i). \end{aligned} \tag{8b}$$

The function[6,28] S_l in (7) is defined by

$$\begin{aligned} S_l &= (1/2^l)(2J)!/(2J - l)! \\ &= J(J - \tfrac{1}{2}) \cdots [J - (l-1)/2] \quad \text{when } l \neq 0. \end{aligned} \tag{9}$$

Single-ion terms are included in (7) as the terms for which either l or l' is zero ($\tilde{O}_{0,0} = 1$ and $S_0 = 1$). Except for the terms for which p is a half integer, the spin Hamiltonian, (7) and (8), includes all possible two-ion and single-ion couplings which affect the magnetic excitations propagating in the c direction and the influence of magnetic ordering on these couplings. The completeness is secured by the use of the tensor operators which span the whole spin space. The correspondence between the Racah operators and the spherical harmonics $Y_{l,m}(\theta,\phi)$ appears from the relation ($m\geq 0$)

$$\frac{1}{S_l}\langle\tilde{O}_{l,m}\rangle=\left(\frac{4\pi}{2l+1}\right)^{1/2}Y_{l,m}(\theta,\phi)=(-1)^m\left(\frac{(l-m)!}{(l+m)!}\right)^{1/2}P_l^m(\cos\theta)e^{im\phi}, \tag{10}$$

when $\langle J_\alpha\rangle=J$, where α is the direction specified by the polar angles θ and ϕ; $P_l^m(\cos\theta)$ are the Legendre polynomials.

The treatment of the general Hamiltonian (7) for obtaining expressions for the spin-wave energies has become more straightforward owing to the systematic investigations of the properties of the Racah operators by Buckmaster *et al.*[5] and by Danielsen and Lindgård.[6] The spin Hamiltonian which we are considering is more complex than the model Hamiltonian generally used for describing the magnetic properties of rare-earth metals[1,28-30] because of the inclusion of general two-ion couplings. The effect of general two-ion interactions on the spin waves in a conically ordered phase has been published elsewhere.[31]

In the case of a basal-plane ferromagnet, such as Tb and Dy, for which $\theta=\frac{1}{2}\pi$ we obtain for the expectation value of the Racah operators

$$(1/S_l)\langle\tilde{O}_{l,m}\rangle=\Gamma_{l,m}e^{im\phi}; \tag{11}$$

when $m\geq 0$ we have

$$\Gamma_{l,m}=(-1)^m[(l-m)!/(l+m)!]^{1/2}P_l^m(0)=\begin{cases}(-1)^{(l+m)/2}\dfrac{[(l+m)!\,(l-m)!]^{1/2}}{(l+m)!!\,(l-m)!!}, & l+m \text{ even}\\ 0, \quad l+m \text{ odd};\end{cases} \tag{12}$$

if m is negative, then

$$\Gamma_{l,m}=(-1)^m\Gamma_{l,-m}. \tag{13}$$

With the purpose of deriving the energies of the spin waves in a transparent way we shall introduce the following expansion of the Racah operators:

$$\tilde{O}_{l,m}\cong\left[1+\delta\theta\frac{\partial}{\partial\theta}+\delta\phi\frac{\partial}{\partial\phi}+\frac{1}{2}\left(\delta\theta\frac{\partial}{\partial\theta}+\delta\phi\frac{\partial}{\partial\phi}\right)^2\right]\langle\tilde{O}_{l,m}\rangle, \tag{14}$$

where $\delta\theta$ and $\delta\phi$ describe the deviations of the spin from alignment along the direction of magnetization (z' direction). When $\theta=\frac{1}{2}\pi$ these deviations are replaced by the following expressions in spin deviation operators a:

$$\begin{aligned}\delta\theta&=(1/J)J_{x'}=(a^\dagger+a)/\sqrt{(2J)},\\ \delta\phi&=(1/J)J_{y'}=i(a^\dagger-a)/\sqrt{(2J)}.\end{aligned} \tag{15}$$

Introducing these expressions in Eq. (14) and calculating the derivatives of the spherical harmonics at $\theta=\frac{1}{2}\pi$, we obtain

$$\frac{1}{S_l}\tilde{O}_{l,m}(J_i)=\left(1-\frac{m}{\sqrt{2J}}(a_i^\dagger-a_i)-\frac{l(l+1)}{2J}a_i^\dagger a_i-\frac{l(l+1)-2m^2}{2J}\frac{1}{2}(a_i^\dagger a_i^\dagger+a_i a_i)\right)\times\Gamma_{l,m}e^{im\phi} \tag{16}$$

if $l+m$ is even. When $l+m$ is odd we find

$$\frac{1}{S_l}\tilde{O}_{l,m}(J_i)=[(l+1)^2-m^2]^{1/2}\times\left(\frac{1}{\sqrt{2J}}(a_i^\dagger+a_i)-\frac{m}{2J}(a_i^\dagger a_i^\dagger-a_i a_i)\right)\times\Gamma_{l+1,m}e^{im\phi}. \tag{17}$$

This result is in agreement with that obtained by a direct transformation[5,6] of the Racah operators to the z'-coordinate system followed by an expansion in spin deviation operators[6] except for some negligible (in the case of a large J value) kinematic effects.[6]

For a hcp structure we have two ions per unit cell, giving rise to acoustic and optical spin-wave branches. The Hamiltonian for a two-sublattice ferromagnet has been diagonalized by Lindgård *et al.*[32] However, as discussed, for example, by Liu,[13] Bose excitations (as the magnons and the phonons) remain doubly degenerate at the hexagonal zone surface AHL. In agreement with this result the magnons described by the Hamiltonian (7) (including terms for which p is a half integer) are doubly degenerate at A. Further, when couplings for which $\mu+m+m'$ is odd are neglected, the spin waves are pure acoustic or optical excitations. This implies that the double-zone representation in the c direction becomes valid, and we shall henceforth use this representation. This simplification allows us to make us of the Fourier transforms defined as

$$K_{\lambda ll'}^{\mu mm'}(\vec{q})=\sum_j K_{\lambda ll'}^{\mu mm'}(\vec{R}_i-\vec{R}_j)e^{i\vec{q}\cdot(\vec{R}_i-\vec{R}_j)}, \tag{18}$$

where the summation is extended over both sublattices. Formally the single-ion terms are included as being independent of $\vec{q}$.

Introducing Eqs. (16)–(18) in the Hamiltonian (7), we obtain

$$\begin{aligned}\mathcal{H}_{MJJ}&=(NJ)E+\sum_q A(\vec{q})a_{\vec{q}}^\dagger a_{\vec{q}}+B(\vec{q})\tfrac{1}{2}(a_{\vec{q}}^\dagger a_{-\vec{q}}^\dagger+a_{\vec{q}}a_{-\vec{q}})\\&=(NJ)E+\sum_q\epsilon(\vec{q})\alpha_{\vec{q}}^\dagger\alpha_{\vec{q}},\end{aligned} \tag{19}$$

where the reduced equilibrium energy (N is the number of ions in the crystal) is given by

$$E=\sum_{p}\sum_{\lambda\mu}\sum_{lm}\sum_{l'm'}\frac{1}{J}K_{\lambda l l'}^{\mu m m'}(0)\times\Gamma_{\lambda,\mu}\Gamma_{l,m}\Gamma_{l',m'}\cos(6p\phi)\delta(\mu+m+m'-6p). \tag{20}$$

The magnon operators ($\alpha_{\vec{q}}^{+}$) are derived by a Bogoliubov transformation of the spin-deviation operators.[28-30,32] The spin-wave energies are then expressed in terms of the two parameters, $A(\vec{q})$ and $B(\vec{q})$. Note that these parameters are both real for a spin wave propagating in the c direction, the energy of which is

$$\varepsilon(\vec{q})=\{[A(\vec{q})+B(\vec{q})][A(\vec{q})-B(\vec{q})]\}^{1/2}. \tag{21}$$

The two Bogoliubov components of the magnon energy at zero wave vector are deduced to be

$$A(0)+B(0)=\sum_{p}\sum_{\lambda\mu}\sum_{lm}\sum_{l'm'}\frac{1}{J}K_{\lambda l l'}^{\mu m m'}(0)\cos(6p\phi)\delta(\mu+m+m'-6p)\times(-[l(l+1)-m^2+l'(l'+1)-m'^2]\Gamma_{\lambda,\mu}\Gamma_{l,m}\Gamma_{l',m'}+2\{[(l+1)^2-m^2][(l'+1)^2-m'^2]\}^{1/2}\Gamma_{\lambda,\mu}\Gamma_{l+1,m}\Gamma_{l'+1,m'}) \tag{22}$$

and

$$A(0)-B(0)=\sum_{p}\sum_{\lambda\mu}\sum_{lm}\sum_{l'm'}\frac{1}{J}K_{\lambda l l'}^{\mu m m'}(0)\cos(6p\phi)\delta(\mu+m+m'-6p)(-1)(m+m')^2\Gamma_{\lambda,\mu}\Gamma_{l,m}\Gamma_{l',m'}. \tag{23}$$

By the inclusion of terms where $\lambda\neq 0$, Eqs. (21)–(23) represent a generalization of the phenomenological macroscopic resonance theory developed by Smit and Belgers[33] (see also Cooper[29]). In the present theory the energy of the uniform spin-wave mode is equal to

$$\varepsilon(0)=(E'_{\theta\theta}E'_{\phi\phi}-E'^{2}_{\theta\phi})^{1/2}. \tag{24}$$

$E'_{\theta\theta}$, etc. denote second derivatives of the equilibrium energy with respect to angle evaluated at the equilibrium position, $\theta=\frac{1}{2}\pi$, where the angular dependences of $\langle\tilde{O}_{\lambda,\mu}\rangle$ are neglected. This follows from

$$E'_{\theta\theta}=A(0)+B(0),\quad E'_{\phi\phi}=A(0)-B(0),\quad E'_{\theta\phi}=0. \tag{25}$$

The dispersion of spin waves propagating in the c direction of a basal-plane ferromagnet is given by

$$A_{\vec{q}}+B_{\vec{q}}=A(\vec{q})+B(\vec{q})-[A(0)+B(0)]=\sum_{p}\sum_{\lambda\mu}\sum_{lm}\sum_{l'm'}\frac{1}{J}[K_{\lambda l l'}^{\mu m m'}(0)-K_{\lambda l l'}^{\mu m m'}(\vec{q})]\cos(6p\phi)\delta(\mu+m+m'-6p)\times(-2)\{[(l+1)^2-m^2][(l'+1)^2-m'^2]\}^{1/2}\Gamma_{\lambda,\mu}\Gamma_{l+1,m}\Gamma_{l'+1,m'} \tag{26}$$

and

$$A_{\vec{q}}-B_{\vec{q}}=A(\vec{q})-B(\vec{q})-[A(0)-B(0)]=\sum_{p}\sum_{\lambda\mu}\sum_{lm}\sum_{l'm'}\frac{1}{J}[K_{\lambda l l'}^{\mu m m'}(0)-K_{\lambda l l'}^{\mu m m'}(\vec{q})]\cos(6p\phi)\delta(\mu+m+m'-6p)2mm'\Gamma_{\lambda,\mu}\Gamma_{l,m}\Gamma_{l',m'}. \tag{27}$$

Owing to the properties of the Legendre polynomials, a simple selection rule appears in the dispersion of the spin waves when $\theta=\frac{1}{2}\pi$. $K_{\lambda l l'}^{\mu m m'}(\vec{q})$ will contribute to $A_{\vec{q}}+B_{\vec{q}}$ if $l+m$ and $l'+m'$ are both odd, and to $A_{\vec{q}}-B_{\vec{q}}$ if $l+m$ and $l'+m'$ are both even and if m and m' are both different from zero. This selection rule is equivalent to the one obtained for a helically ordered structure.[31]

For spin waves propagating in directions other than the highly symmetric c direction, the condition (8) is not necessarily fulfilled. Terms in which $\mu+m+m'\neq 6p$ give rise to a lifting of the degeneracy along the K-H edge in Tb, as discussed by Lindgård and Houmann[34] in a paper presenting the first indication of anisotropic two-ion couplings in Tb other than the normal magnetic dipole interaction. Only in special cases such as this, and the conically ordered spin system (Er[31,35]), is it possible to detect anisotropic two-ion couplings simply from studies of the dispersion relation. To detect such couplings in a ferromagnetic spin system we need more information, which may be obtained from the interaction of the spins with other systems such as the lattice or thermal neutrons (the scattering cross section), or from the influence of external disturbances on the spin-wave spectrum. The application of an external magnetic field (or a uniaxial stress) affects the spin-wave energies in a way which makes a distinction between $A(\vec{q})+B(\vec{q})$ and $A(\vec{q})-B(\vec{q})$ possible.

In the presence of an external field the Zeeman Hamiltonian

$$\mathcal{H}_Z = -g\mu_B \sum_i \vec{J}_i \cdot \vec{H} \tag{28}$$

will contribute only to the $A(\vec{q})$ part; thus when the field is parallel with the direction of magnetization, Eq. (21) is replaced by

$$\varepsilon(q) = \{[A(\vec{q}) + B(\vec{q}) + g\mu_B H][A(\vec{q}) - B(\vec{q}) + g\mu_B H]\}^{1/2}, \tag{29}$$

where we have neglected a possible change of the strength of couplings in (7) with magnetic field. As long as the external field is much smaller than the exchange field ($H_{Ex} \simeq 20$ MOe in Tb) and the temperature is close to zero, the effect of the external field on the exchange interaction (by polarizing the conduction electrons) can be taken into account by defining an effective g value[36]

$$g_{eff} = 4\pi M(0)/N\mu_B J = 1.038 g, \tag{30}$$

where $M(0)$ is the magnetization at zero temperature.[37,38]

The relations deduced above are only strictly valid at zero temperature. As the temperature is raised the relative magnetization defined as

$$\sigma(T) = M(T)/M(0) \tag{31}$$

decreases. Callen and Callen[39] have developed a theory in which the spin correlation functions, $\langle \tilde{O}_{l,m}(J_i)\rangle$ are expressed as functions of σ. The effective renormalization of the magnon energies due to magnon-magnon interaction is then obtained from this theory, following Cooper,[29] by dividing the spin correlation functions in the expression for the magnon energies by σ [this is obtained as a generalization of the result (24), which at finite temperature should be divided by σ]. The σ dependence of these functions as deduced by Callen and Callen should follow the classical Zener power law at low temperatures, which implies that

$$(1/J) K_{\lambda l l'}^{\mu m m'}(\vec{q}) \propto \sigma^{\gamma(\lambda, l, l')-1}, \tag{32}$$

$$\tfrac{1}{2}[\lambda(\lambda+1) + l(l+1) + l'(l'+1)] \leq \gamma(\lambda, l, l') \leq \tfrac{1}{2}[\lambda(\lambda+1) + (l+l')(l+l'+1)]. \tag{33}$$

If l and l' are both different from zero, $\gamma(\lambda, l, l')$ depends, within the limits given, on the actual correlation range of the coupling. Brooks *et al.*[28,40] have considered the effects of strong anisotropy on this temperature law and have obtained first-order correction in the case of noncylindrical symmetry (as in Tb and Dy). Lindgård and Danielsen[41] have treated the same problem in a Hartree-Fock approximation. They found that the power-law dependence of the spin correlation functions, Eq. (32), should still be valid; however, the relation (33) is modified in a way which differs somewhat from that deduced by Brooks. In this context the relation (32) is the most important, because the dependence of a certain spin correlation function on the intensive variables (H and T) is then described by one parameter γ instead of two. The relation between the field and temperature dependence is determined by (32) if the relative magnetization σ is known as function of H and T. The condition (33) is here only used as an indicative relation between λ, l, and l' and the γ deduced from experiments.

Hegland *et al.*[37] have determined the magnetization in Tb as a function of temperature. Their measurements also give some idea of the magnitude of $d\sigma/dH$. The molecular-field value for $d\sigma/dH$ is[39]

$$d\sigma/dH \cong g\mu_B J(1-\sigma)/k_B T_N, \tag{34}$$

where T_N is the Néel temperature (225 K in Tb). This value is comparable to that deduced from the magnetization measurements,[37] and it agrees with the one deduced from the observed forced magnetostriction in Tb,[42] as shown in III. At finite temperatures ($\sigma<1$) the application of an external field will give rise to an increase of the relative magnetization. The field dependence of σ connected with the implicit σ dependence of $A(\vec{q})$ and $B(\vec{q})$ as given by (32) introduces a correction to the energy expression (29); so the field dependence of the square of the magnon energies at finite temperature is (in the limit of small fields)

$$\frac{d\varepsilon^2(\vec{q})}{dH} = 2g\mu_B A(\vec{q}) + \frac{\partial \varepsilon^2(\vec{q})}{\partial\sigma}\,\frac{d\sigma}{dH}. \tag{35}$$

Thus the field dependence of the magnon energies is simply related to $A(\vec{q})$, which, combined with the absolute value of the energies, leads to a determination of $A(\vec{q})+B(\vec{q})$ and $A(\vec{q})-B(\vec{q})$.

IV. EXPERIMENTAL DETERMINATION OF ANISOTROPY

The spin-wave dispersion relations in Tb and in Tb-Ho alloys have been extensively studied by inelastic-neutron-scattering experiments.[43] In these experiments the dispersion relations along the symmetry lines in the Brillouin zone have been determined over a wide temperature range. The lifetime and the energies of spin waves propagating in the c direction have been studied both in the spiral (216–225 K) and in the ferromagnetic (< 216 K) phases. A recent review can be found in the article by Mackintosh and Bjerrum Møller.[44]

Common to the interpretation of the experimental results in these previous publications on Tb, and to other works concerning the magnetic properties of the heavy-rare-earth metals, is the assumption of an isotropic two-ion coupling as given by Eq. (1). However, the interpretation[31,35] of the spin-wave measurements of Nicklow *et al.*[35] in the conical magnetic phase of Er showed unambiguously that anisotropic two-ion couplings are important in determining the magnetic properties of Er. These measurements prompted us to investigate the validity of the assumption of isotropy in Tb. As shown in Sec. III, a way to proceed is to measure the field dependence of the magnon energies. This method has been used for obtaining the magnetic anisotropy at zero wave vector in[45] Tb–10-at.%-Ho and in pure Tb (III).

The energy of spin waves propagating in the c direction of Tb was studied by inelastic neutron scattering. The energies were measured as a function of magnetic field applied along both the easy and hard directions in the basal plane at three different temperatures (4.2, 53, and 134 K, corresponding to a relative magnetization[37] of 1, 0.971, and 0.832, respectively). An external field of up to 100 kG could be applied.

A selection of the experimental results at 53 K for different $\vec{q}$ values is shown in Fig. 1. The terms quadratic in field, $(g\mu_B H_i)^2$ and $[g\mu_B(H_i-H_c)]^2$ for H parallel to an easy and a hard axis, respectively, are subtracted from the square of the magnon energies shown in the figure. H_c is the critical field necessary to pull the moments into alignment parallel to a field applied in a hard direction (see III). The energies squared should then increase linearily with the internal field H_i, according to the Eqs. (29) and (35).

The observed coupling between the magnons and the transverse phonons propagating in the c direction perturbs the field dependence of the magnon energies. The magnons interact both with the acoustic and optical transverse phonons. The abnormal acoustic-optical coupling raises a severe problem, as it violates the selection rules deduced in II for a basal-plane ferromagnet. The acoustic magnons are observed to be coupled with the optical transverse phonons which have their polarization vector parallel with a b axis, whereas general symmetry arguments predict that the optical transverse phonons which may couple to the acoustic magnons are those polarized parallel to an a axis. The occurrence of an acoustic-optical interaction requires the spin and space variables to be directly mixed, and Liu[13] has proposed a mechanism which involves the spin-orbit coupling of the conduction electrons. In II we find that this mechanism, besides making an acoustic-optical coupling possible as proposed by Liu, also introduces new

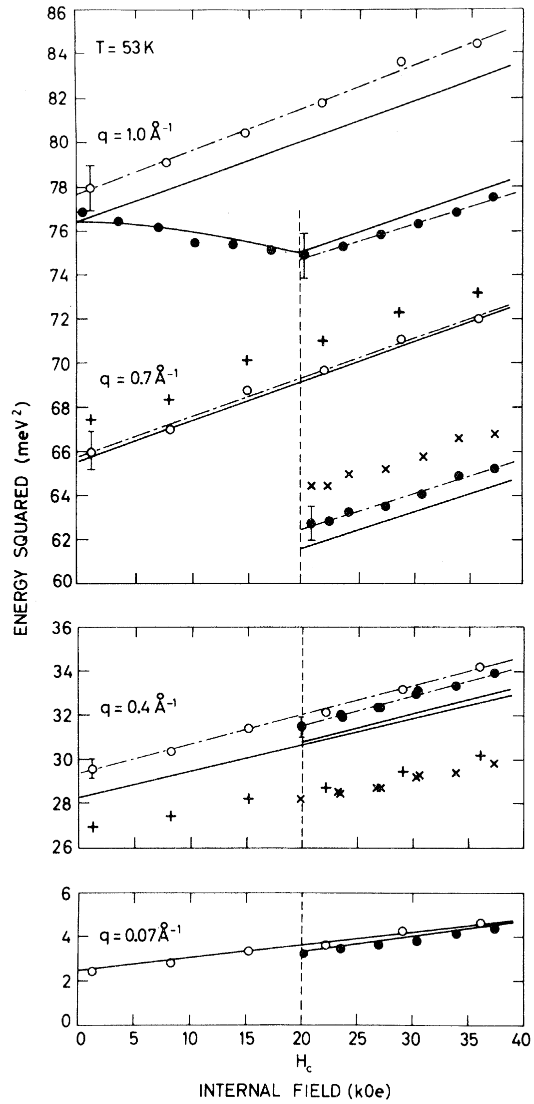

FIG. 1. Dependence of the square of the magnon energies for different $\vec{q}$ values in the c direction on internal field in Tb at 53 K. The energies have been corrected for the terms quadratic in field as described in the text. The symbols + and 0 represent results for the field in easy direction, and × and ● the hard direction. The experimental results, + and ×, were corrected for the influence of the magnon-phonon interaction, and the circles are the energies of the unperturbed magnons obtained after introducing this correction, which is negligible at $\vec{q}$ values of 0.07 and 1.00 Å^{-1}. The dashed lines illustrate the linear behavior of the energy squared as a function of field, and the solid lines are the final results of the least-squares analysis.

selection rules to first order in the spin-orbit parameter $\langle \lambda_{s\text{-}o} \rangle$. The spin-orbit coupling implies that the spin-up and spin-down states of the conduction electrons are mixed, which corresponds to a deviation between the direction of magnetization and the direction in which the conduction electrons are polarized (Tb cannot then be considered as a simple ferromagnet). The equations of motion of the total magnon-phonon system are considered in detail in II. Both the normal magnon-phonon interaction and the abnormal acoustic-optical coupling vanish at Γ ($q = 0$ and $2\pi/c$) and are negligible when q is 0.07 and 1.00 Å^{-1}, as is apparent from Fig. 1, where we have shown the square of the perturbed magnon energies and the results when the energies have been corrected for the couplings. The way in which this correction has been carried out is described in II. With the exception of the results shown in Fig. 1 all the experimental results which appear in this article have been corrected for the direct coupling between the magnons and the phonons.

The uncertainties introduced by the correction for magnon-phonon interactions are not included in the standard deviations of the experimental results in Figs. 1 and 2. In a neutron experiment a relatively small change of energies can be determined more accurately than given by the absolute uncertainties of the energies, which is the reason for the striking linear behavior of the energies squared (the dashed lines) when compared with the (absolute) standard deviations shown in Fig. 1. In Fig. 2 we show the dispersion relations in the c direction at zero field, 2(a), and at a field of H_c applied in a hard direction, 2(b), as a function of temperature.

The initial slope $\alpha(\vec{q})$ of the square of the magnon energies as a function of the reduced field $g_{\text{eff}}\mu_B H_i$,

$$\alpha(\vec{q}) = \frac{1}{g_{\text{eff}}\mu_B} \frac{d\epsilon^2(\vec{q})}{dH} \quad \text{when } H_i \to \begin{cases} 0, & H \neq b \text{ axis} \\ H_c, & H \neq a \text{ axis} \end{cases} \tag{36}$$

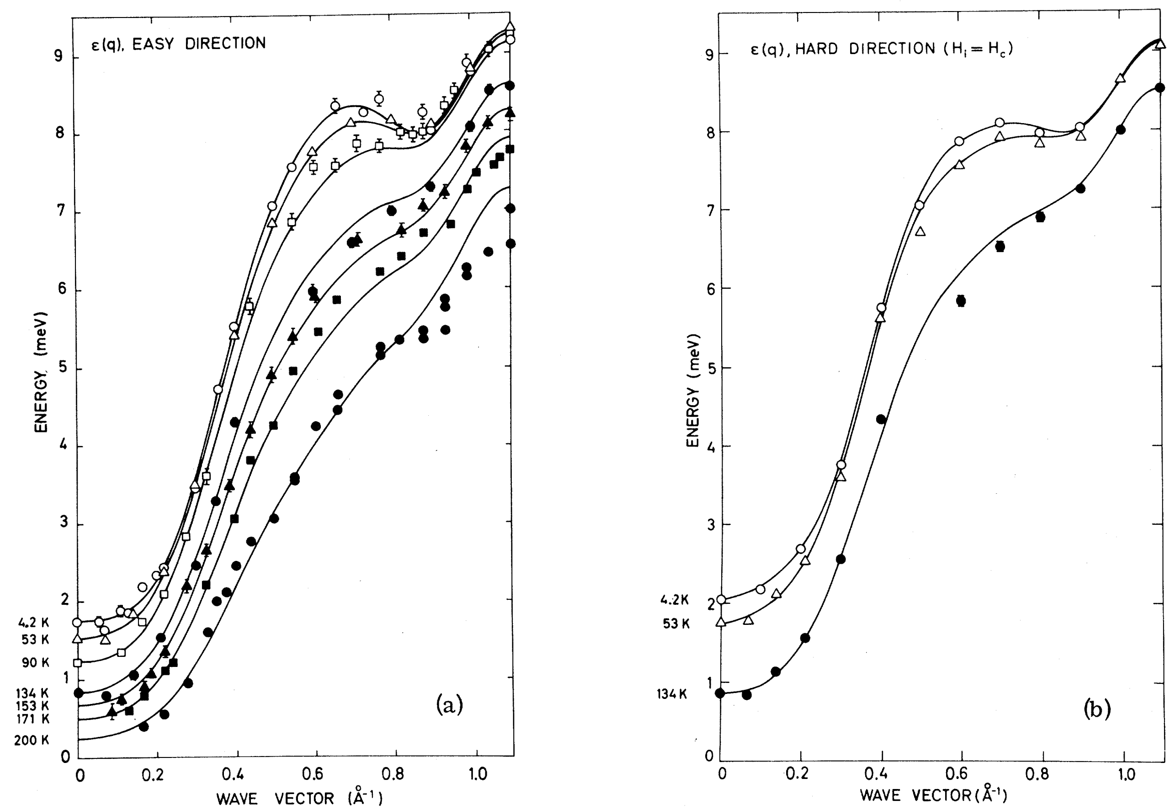

FIG. 2. Dispersion relations for the unperturbed spin waves propagating in the c direction of Tb as functions of temperature, (a) when the magnetization is along an easy axis, and (b) when it is along a hard axis. The uncertainties introduced by the correction for magnon-phonon interaction, which is largest for $\vec{q}$ around 0.55 Å^{-1}, are not included in the standard deviations of the experimental results. The standard deviations of the results at 171 and 200 K in (a) are not shown.

is shown in Fig. 3. Here we use the effective g value defined in Eq. (30). At low temperature $\alpha(\vec{q})$ is simply equal to $2A(\vec{q})$, whereas the temperature dependence of the magnetization introduces additional contributions at finite temperatures, Eq. (35). This is illustrated in Fig. 3(c), where the dashed line indicates the final result for $2A(\vec{q})$; as seen in the figure, the σ dependence of the crystal field and exchange parameters makes a contribution of about 25% to the slope $\alpha(\vec{q})$, when σ is equal to 0.832.

At low temperature (4.2 K) we obtain immediately $A_{\vec{q}} \pm B_{\vec{q}}$ [defined in (26) and (27)] from

$$A_{\vec{q}} \pm B_{\vec{q}} = \tfrac{1}{2}\alpha(\vec{q}) \pm \tfrac{1}{2}[\alpha^2(\vec{q}) - 4\epsilon^2(\vec{q})]^{1/2} - [A(0) \pm B(0)], \tag{37}$$

where the spin-wave parameters at zero wave vector, $A(0) \pm B(0)$, are known from the field experiment described in III. A measurement of $\epsilon(\vec{q})$ and $\alpha(\vec{q})$ in both the cases in which the magnetization is along an easy and a hard direction determines four spin-wave parameters. At 4.2 K those four parameters are $A_{\vec{q}} \pm B_{\vec{q}}$ at $\phi = 0$ and $\phi = \frac{1}{2}\pi$, which are shown in Fig. 4. The general expressions (26) and (27) offer no possibilities for a reduction of this number. In the case of an isotropic two-ion coupling (1), $B_{\vec{q}}$ should be zero and $A_{\vec{q}}$ should be independent of ϕ and should, within a random-phase approximation, renormalize as σ:

$$\{A_{\vec{q}}\}_{\rm iso} = J[\mathcal{J}(0) - \mathcal{J}(\vec{q})]\sigma; \quad \{B_{\vec{q}}\}_{\rm iso} = 0. \tag{38}$$

This relation is not fulfilled in the case of Tb, as is apparent from Fig. 4; $B_{\vec{q}} \approx \frac{1}{2}A_q$ when the magnetization is along an easy axis, and the parameters depend on ϕ. The terms which introduce a ϕ dependence of $A_{\vec{q}} \pm B_{\vec{q}}$ arise from couplings of high rank $(\lambda + l + l' \geq 6)$ which according to (33) should renormalize quite rapidly $(\gamma \geq 9)$. If we neglect terms proportional to $\cos 12\phi$ and $\cos 18\phi$, which presumably are small, the ϕ-dependent terms are easily extracted. Because anisotropic couplings in general contribute differently to $A_{\vec{q}} + B_{\vec{q}}$ and $A_{\vec{q}} - B_{\vec{q}}$, see Eqs. (26) and (27), the isotropic part of the two-ion coupling is not directly related to the ϕ-independent part of $A_{\vec{q}}$. However, the experimental results obtained at finite temperatures can give some indications of the magnitude of $J[\mathcal{J}(0) - \mathcal{J}(\vec{q})]$ if most of the anisotropic contributions renormalize faster than σ. A more definitive result could be obtained if it were possible to create a significant magnetic moment along the c axis.

In order to deduce $A_{\vec{q}} \pm B_{\vec{q}}$ at finite temperatures we have to solve Eqs. (21) and (35) self-consistently. For this purpose we parametrized $A_{\vec{q}} \pm B_{\vec{q}}$ in several different ways, and obtained clearly the best least-squares fit by using the parameters defined as

$$A_{\vec{q}} + B_{\vec{q}} = \mathcal{I}(\vec{q})\sigma^{j(\vec{q})} + \mathcal{K}(\vec{q})\sigma^{k(\vec{q})} - \mathcal{C}(\vec{q})\sigma^{k(\vec{q})}\cos 6\phi \tag{39a}$$

and

$$A_{\vec{q}} - B_{\vec{q}} = \mathcal{I}(\vec{q})\sigma^{j(\vec{q})} - \mathcal{K}(\vec{q})\sigma^{k(\vec{q})} - \mathcal{D}(\vec{q})\sigma^{k(\vec{q})}\cos 6\phi, \tag{39b}$$

where ϕ is the angle between the direction of magnetization and the a axis (hard direction) and by definition all the coupling parameters vanish identically at zero wave vector. The experimental results allow us to introduce not more than four coupling parameters. These four parameters are then $\vec{q}$-dependent linear combinations of terms which may depend differently on σ (according to their rank, $\lambda + l + l'$). Within the quite narrow range of σ $(1 \geq \sigma \geq 0.8)$ which we consider, we may hope that the σ dependence of the four parameters are satisfactorily accounted for by introducing four effective exponents. These exponents cannot then be expected to be independent of $\vec{q}$.

The way in which the coupling parameters are defined by (39) turned out to be the most appropriate one. One of the advantages obtained when the ϕ-independent part of $A_{\vec{q}} \pm B_{\vec{q}}$ is parametrized as in (39) is that there was no experimental evidence for distinguishing between the σ dependence of the three anisotropy parameters $\mathcal{K}(\vec{q})$, $\mathcal{C}(\vec{q})$, and $\mathcal{D}(\vec{q})$, and hence the number of parameters can be reduced by two. The distinction made between $\mathcal{I}(\vec{q})$ and $\mathcal{K}(\vec{q})$ is justified by the final result that $\mathcal{I}(\vec{q}) > (>) \mathcal{K}(\vec{q})$, shown in Fig. 5, and especially by the large difference in the σ dependence obtained, $j(\vec{q}) \ll k(\vec{q})$, as shown in Fig. 7. The result for $j(\vec{q})$, which is found to lie between 0 and 2, implies that the behavior of $\mathcal{I}(\vec{q})$ is rather close to that expected for the isotropic part of the two-ion coupling.

The final fit was obtained by expressing the parameters in (39) in terms of cosine series with interplanar coupling constants as coefficients, e.g.,

$$\mathcal{I}(\vec{q}) = \sum_n \mathcal{I}_n[1 - \cos(\zeta n\pi)], \tag{40}$$

where $\zeta = qc/2\pi$. The values obtained for the interplanar coupling constants $\mathcal{I}_n$, $\mathcal{K}_n$, $\mathcal{C}_n$, and $\mathcal{D}_n$ are given in Table I together with their uncertainties. The corresponding curves are the solid lines in Figs. 5 and 6. The magnetization exponents $j(\vec{q})$ and $k(\vec{q})$ are expanded in cosine series as

$$j(\vec{q}) = j_0 + \sum_n j_n[1 - \cos(\zeta n\pi)], \tag{41}$$

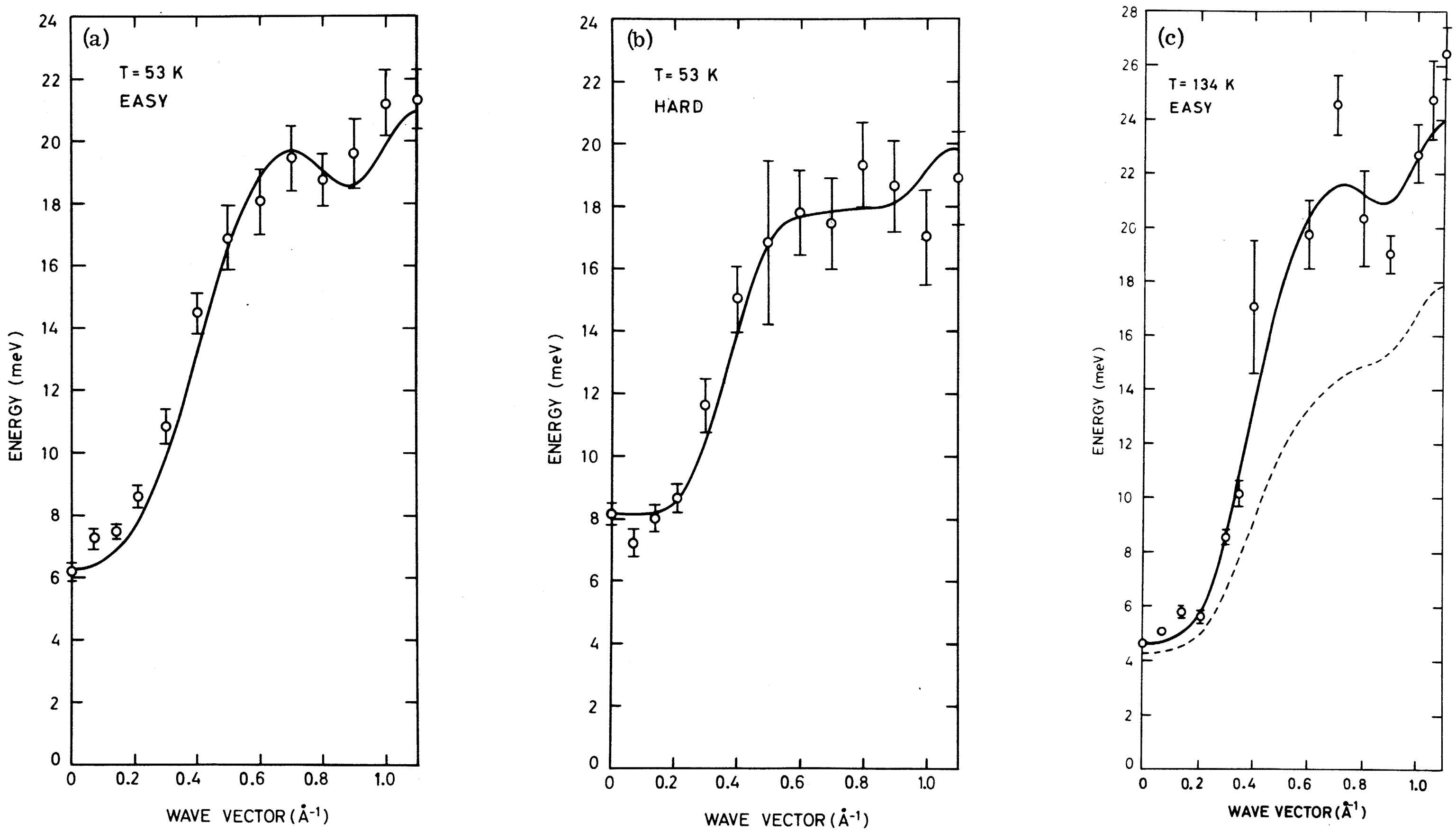

FIG. 3. Initial slope $\alpha(\vec{q})$ of the squared magnon energies in Tb as a function of the reduced field, $g_{eff}\mu_B H_i$, which is equal to $2A(\vec{q})$ at zero temperature. $\alpha(\vec{q})$ is shown for the field applied along both the (a) easy and (b) hard directions at 53 K, which correspond to the slopes of the dashed lines in Fig. 1. The difference between $\alpha(\vec{q})$ and $2A(\vec{q})$ may be substantial at higher temperatures because of the field dependence of the magnetization, and in (c) the experimental results for $\alpha(\vec{q})$ at 134 K are compared with the calculated values of $2A(\vec{q})$, which are represented by the dashed line.

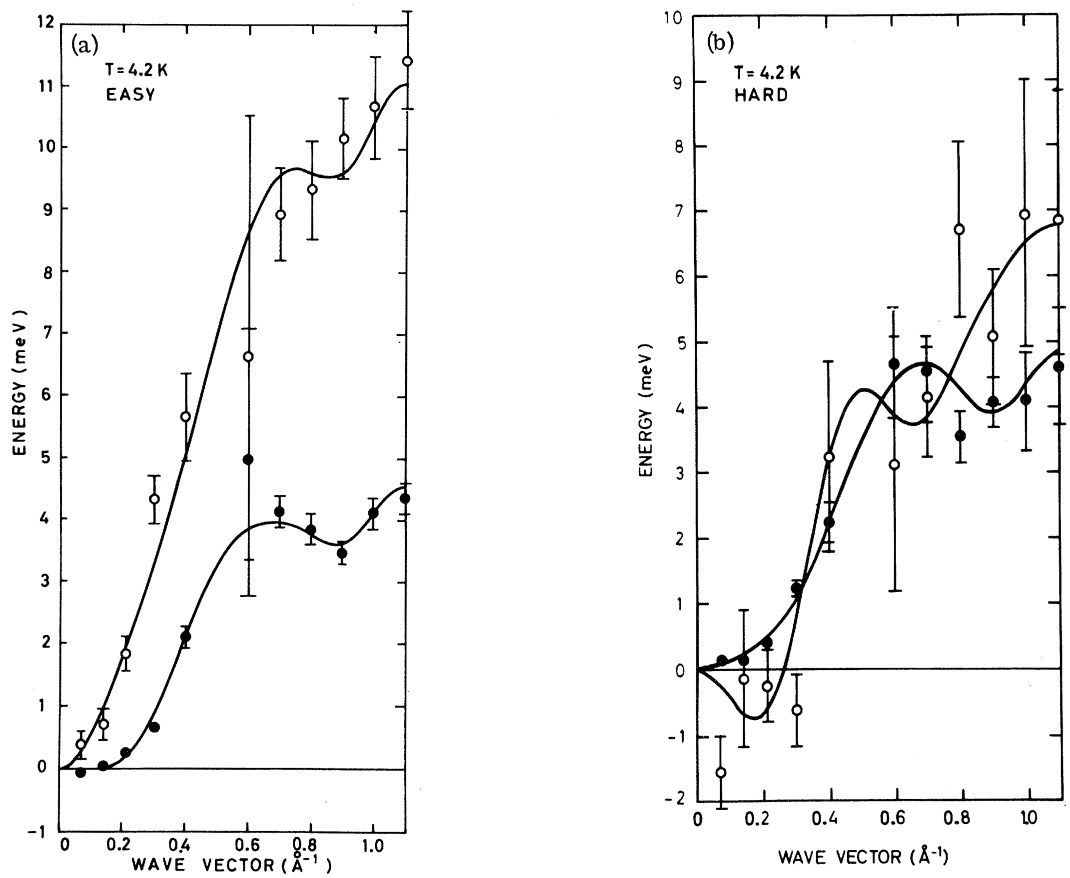

FIG. 4. $\vec{q}$ dependence of the two components, $A_{\vec{q}}+B_{\vec{q}}$ and $A_{\vec{q}}-B_{\vec{q}}$, of the magnon energies in the c direction of Tb at 4.2 K (open and closed symbols, respectively). The effective $\vec{q}$-dependent anisotropy is given by half the difference between the two energy components, $B_{\vec{q}}$, which is deduced to be of an appreciable magnitude when the magnetization is along an easy axis, (a), whereas $B_{\vec{q}}$ is quite small in the hard axis configuration, (b).

the coefficients of which are given in Table II and reproduced in Fig. 7.

The four two-ion coupling parameters in conjunction with the two corresponding magnetization exponents, which are shown in Figs. 5–7, reproduce satisfactorily the field and temperature dependence of the magnon energies. The final least-squares fit appears as solid lines on all the figures shown. The two high-temperature dispersion relations at 171 and 200 K in Fig. 2(a) were not included in the least-squares fitting because of an expected breakdown of the simple effective magnetization dependences of the parameters (200 K corresponds to $\sigma = 0.60$). Nevertheless, an extrapolation of the coupling parameters up to these temperatures produces dispersion relations which agree very well with the measured magnon energies. This agreement indicates that the low-temperature parameters are the relevant ones also at higher temperatures, and at temperatures above ~150 K the anisotropy parameters $\mathcal{K}(\vec{q})$, $\mathcal{C}(\vec{q})$, and $\mathcal{D}(\vec{q})$ are all negligible in comparison with $\mathcal{J}(\vec{q})$, which allows us to conclude that the two-ion coupling in Tb is effectively isotropic above this temperature ($B_{\vec{q}} \approx 0$). $j(\vec{q})$ in Fig. 7 deviates somewhat from 1 (except when $q \approx 0.55$ Å^{-1}), which suggest that $\mathcal{J}(\vec{q})$ contains anisotropic contributions. If we assume that the isotropic part renormalizes as σ, it is possible to make an estimate of its magnitude by using the dispersion relations determined at the higher temperatures where $A_{\vec{q}} \gg B_{\vec{q}}$. This estimate is shown in Fig. 5 as the dashed line. Because of the assumptions involved we shall only claim that

$$\mathcal{J}(\vec{q}) \approx J[\mathcal{J}(0) - \mathcal{J}(\vec{q})] \qquad (42)$$

and consider the relatively small deviation of $\mathcal{J}(\vec{q})$ from the dashed line in Fig. 5 to be a measure of the uncertainty with which the isotropic part of the two-ion coupling in Tb has been determined.

In Fig. 4 was shown $A_{\vec{q}} \pm B_{\vec{q}}$ at 4.2 K deduced for the two cases where the moments are either along

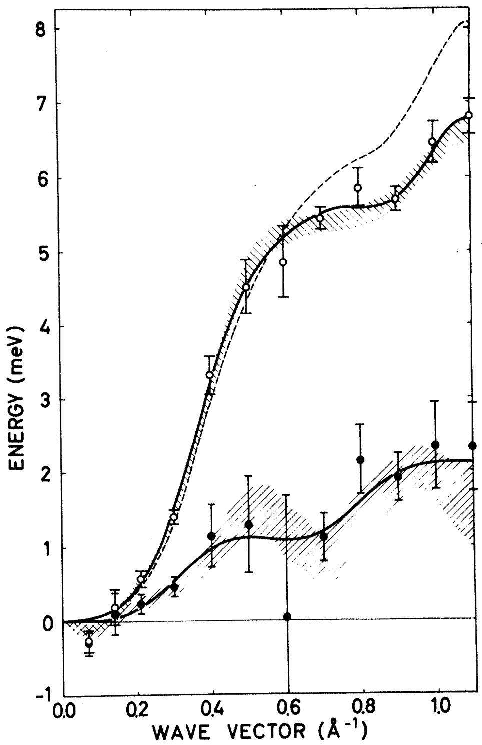

FIG. 5. Coupling parameters $\mathcal{J}(\vec{q})$ and $\mathcal{K}(\vec{q})$ for Tb, as defined by Eq. (39), represented by open and closed circles, respectively. $\mathcal{J}(\vec{q})$ may be considered to be a fair representation of the isotropic two-ion contribution $J[\mathcal{J}(0)-\mathcal{J}(\vec{q})]$. An estimate of this isotropic part, which is discussed in the text, is shown by the dashed line. $\mathcal{K}(\vec{q})$ represents the ϕ-independent anisotropy. All coupling parameters are the spin-wave parameters at zero temperature. The circles are the results deduced by a least-squares fitting to all experimental results obtained at a certain $\vec{q}$ value, most of which are shown in Figs. 2 and 3. The statistical scatter of the experimental results, $\varepsilon(\vec{q})$ and $\alpha(\vec{q})$, and the uncertainties introduced by the magnon-phonon interaction were smoothed by fitting a Fourier expansion to the results. The standard deviations obtained by the least-squares analysis of the Fourier results are presented as the cross-hatched areas. These areas then show the probable regions in which the parameters lie. The best fit of the two coupling parameters which was obtained is represented by the solid lines. This final least-squares fit appears as solid lines on all the figures shown.

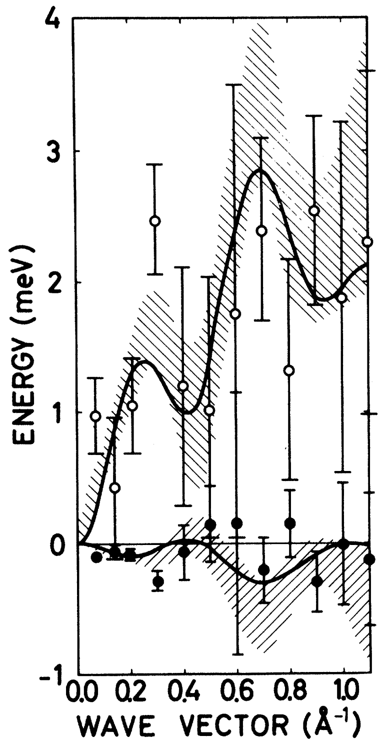

FIG. 6. Basal-plane anisotropy parameters $\mathcal{C}(\vec{q})$ (open symbols) and $\mathcal{D}(\vec{q})$ (closed symbols) representing the dependence at zero temperature of the two-ion anisotropy in Tb on the orientation of the moments in the basal plane, see Eq. (39). The way in which the parameters were derived and the meaning of the signatures used are explained in the caption to Fig. 5.

an easy axis, 4(a), or along a hard axis, 4(b). The variation of the neutron cross section with the wave vector is directly related to these parameters. In the case where $B(\vec{q})$ is real and the scattering vector $\vec{\kappa}$ is along the c direction, the $\vec{q}$ dependence of the neutron cross section is given by [32,44]

$$\frac{d^2\sigma}{d\Omega\,dE} \propto \frac{k'}{k_0} F^2(\vec{\kappa}) \sum_{\tau} \frac{A(\vec{q})+B(\vec{q})}{\varepsilon(\vec{q})} (n_{\vec{q}}+1) \times \delta(\vec{\kappa}-\vec{q}-\vec{\tau})\delta(E-\varepsilon(\vec{q})) \,. \tag{43}$$

$\vec{k}_0$ and $\vec{k}'$ are the wave vectors of the incident and scattered neutrons, respectively; $F(\vec{\kappa})$ is the magnetic form factor[46]; $\vec{\tau}$ is a reciprocal-lattice vector; and $n_{\vec{q}}$ is the boson distribution function. The integrated intensities of the neutron groups obtained experimentally by monochromator scans (k' constant) are compared with the intensities deduced from (43) in Fig. 8. The agreement is quite good (the solid line); however, because of the arbitrariness of the absolute intensity scale, the comparison in Fig. 8 is not very sensitive to the $\vec{q}$-dependent anisotropy, as is illustrated by the dashed line for which $B(\vec{q})$ is taken to be independent of $\vec{q}$ [equal to $B(0)$].

In a preliminary interpretation of the experimental results[47] the magnon-phonon interaction

TABLE I. Interplanar coupling constants for Tb as defined in the text. Here and in Table II $s(\ )$ denotes the standard deviation of the corresponding parameter. All values are in meV.

n	$\mathcal{J}_n$	$s(\mathcal{J}_n)$	$\mathcal{K}_n$	$s(\mathcal{K}_n)$	$\mathcal{C}_n$	$s(\mathcal{C}_n)$	$\mathcal{D}_n$	$s(\mathcal{D}_n)$
1	3.203	0.048	0.980	0.098	0.841	0.229	−0.041	0.078
2	0.846	0.077	0.022	0.140	0.403	0.290	−0.061	0.097
3	0.144	0.052	0.193	0.108	−0.220	0.242	0.097	0.080
4	−0.388	0.075	−0.008	0.124	0.168	0.260	−0.029	0.087
5	−0.018	0.059	−0.115	0.087	0.444	0.206	−0.062	0.053
6	−0.084	0.061						
7	0.064	0.061						

and the field dependence of σ were neglected. These approximations combined with the ambiguity in sign in front of square root, $B(\vec{q})$, in Eq. (37) [when $B(\vec{q})$ becomes close to zero] led to coupling parameters which differ substantially from those deduced in the present analysis.

V. DISCUSSION

The two-ion spin Hamiltonian in Tb is dominated by the coupling parameter, $\mathcal{J}(\vec{q})$, which is presumably isotropic and renormalizes approximately as σ. The exchange parameter deduced from the spin-wave measurements in Gd[48] may include anisotropic contributions originating in the spin-orbit coupling of the conduction electrons (ii). Neither is the parameter $J[\mathcal{J}(0) - \mathcal{J}(\vec{q})]$ in Er derived by Jensen[31] from spin-wave measurements[35] necessarily isotropic. In spite of the uncertainties associated with the two-ion couplings deduced from experiment, the appropriately scaled function $[\mathcal{J}(\vec{q}) - \mathcal{J}(0)]/(g-1)^2$ is similar, both in magnitude and the general $\vec{q}$ dependence, for these heavy-rare-earth metals, as shown in Fig. 9. The differences which appear are probably mainly due to a variation of the properties of the band electrons,[14-16] as discussed by Lindgård and Liu.[16] In Er the maximum is essential for stabilizing the periodic magnetic ordering. The apparent agreement between the scaled isotropic part of the two-ion spin Hamiltonian for the three heavy-rare-earth metals Gd, Tb, and Er supports the RKKY theory in emphasizing the importance of the indirect exchange interaction via the conduction electrons, Eq. (1).

At zero temperature, the two-ion anisotropy parameters $\mathcal{K}(\vec{q})$ and $\mathcal{C}(\vec{q})$ are of the order of $\frac{1}{3}$ of the isotropic coupling, whereas $\mathcal{D}(\vec{q})$ is almost negligible. They all renormalize quite rapidly, as σ^{11} to σ^{27} (depending on $\vec{q}$), which implies that $\mathcal{J}(\vec{q})$ is the only coupling parameter of significance in the spin-wave Hamiltonian at higher temperatures (above 150 K). The magnitude of the anisotropic part of the two-ion coupling in Er[31] is found to be slightly larger than the magnitude of the isotropic part at low temperatures. Recent measurements on Dy[49] (similar to those performed on Tb) show Dy as lying between Tb and Er with respect to the anisotropy. This indication of an increase of the importance of anisotropic two-ion couplings in the heavy rare-earths with atomic number, and hence with L, may

TABLE II. Fourier components of the magnetization exponents which implicitly describe the temperature dependence of the two-ion coupling parameters in Tb according to the text.

n	j_n	$s(j_n)$	k_n	$s(k_n)$
0	2.000	0.202	27.48	4.66
1	−0.960	0.122	−7.72	2.27
2			−2.99	1.46

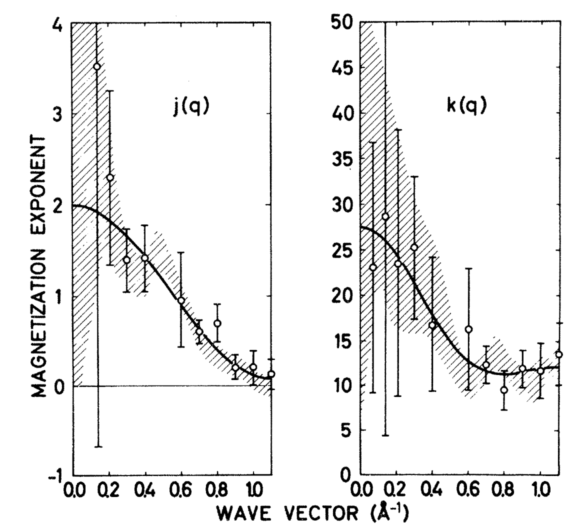

FIG. 7. $j(\vec{q})$ and $k(\vec{q})$ obtained by the least-squares analysis as described in the caption to Fig. 5. $j(\vec{q})$ is the power dependence of $\mathcal{J}(\vec{q})$ with relative magnetization, and $k(\vec{q})$ is the exponent for the anisotropy parameters $\mathcal{K}(\vec{q})$, $\mathcal{C}(\vec{q})$, and $\mathcal{D}(\vec{q})$.

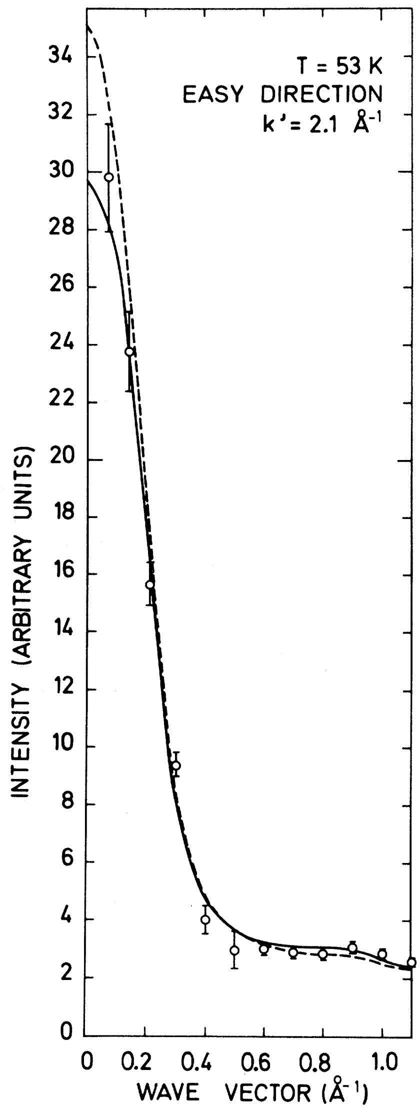

FIG. 8. Integrated intensities of the neutron groups arising from magnon creation in a monochromator scan ($k'=2.1$ Å^{-1}) in Tb at 53 K when the moments are along an easy axis. The solid line represents the $\vec{q}$ dependence of the neutron scattering cross section deduced from the final results [Eq. (43)], whereas the dashed line shows the behavior if the $\vec{q}$-dependent anisotropy is neglected.

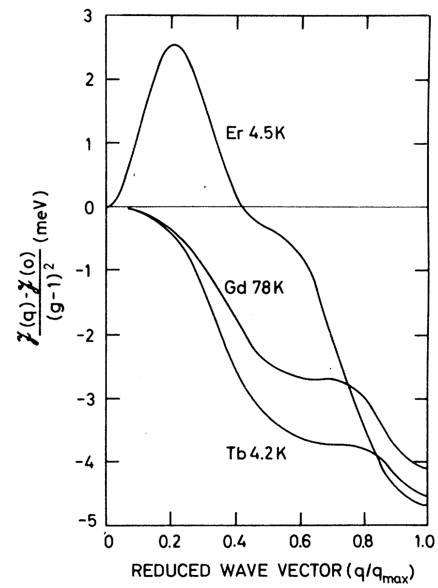

FIG. 9. Exchange function $[\mathcal{J}(\vec{q})-\mathcal{J}(0)]/(g-1)^2$ in the c direction for the heavy rare-earth metals Gd, Tb, and Er.

serve as a guide in investigations of the mechanisms responsible for the observed anisotropy. Measurements of the dispersion of the crystal-field levels in Pr [34,50] show that anisotropic two-ion interactions are also important in the light-rare-earth metals.

The identification of the microscopic mechanisms from which the $\vec{q}$-dependent anisotropies in the rare-earth metals originate requires both further experiments and detailed theoretical calculations. However, some qualitative arguments may be suggested, based on the results obtained for Tb. Birgeneau and Kjems[51] have suggested that the large phonon-induced quadropole interactions observed in the rare-earth vanadates[22] may be of equal importance in the metals (viii). It is known that magnetoelastic effects (II, III, and Ref. 52) are important in Tb, and there is therefore the possibility of large phonon-induced couplings. The correction of magnon energies for the perturbation due to the interaction with the transverse phonons propagating in the c direction takes the most important contributions into account. As discussed in II, there remain at low temperature the higher-order zero-point contributions arising from the indirect interactions transmitted by a simultaneous emission and/or absorption of a magnon and a phonon. These terms are similar to those due to magnon-magnon interactions and are presumably small, as they do not seem to be present at zero wave vector (III), and they cannot explain the strong ϕ dependence of the anisotropy $\mathfrak{C}(\vec{q})$. The most important phonon-magnon induced coupling which contributes to $A_{\vec{q}}+B_{\vec{q}}$ is the normal ϵ-mode coupling, which also appears as the direct magnon-phonon interaction in the c direction (l and l' both even and m and m' both odd), and this coupling does not show any ϕ dependence (see II). The γ-mode interaction, giving rise to the ϕ-dependent distortion of the hexagonal symmetry of the basal plane, does not contribute to $A_{\vec{q}}+B_{\vec{q}}$. The strongest magnon-phonon interaction which has been observed in Tb is the acoustic-optical interaction (II), and the origin of this coupling is not the Coulomb interaction but the exchange interaction via spin-orbit coupled states of the conduction electrons, (ii), as proposed by Liu.[13] This magnon-phonon coupling has also been observed in Dy,[53] and it explains the occurrence of an energy gap in the spin-wave spectrum of Er[35] (see II). The importance of the spin-orbit-coupled states in Tb is further supported by the indication of an interaction between the acoustic and optical magnons. Such an interaction, which we have neglected in our analysis, may be present in the case of an a-axis magnet and arises from the terms in (7) for which p in (8a) is a half integer ($p=\frac{1}{2}$ introduces a coupling proportional to $\cos 3\phi$). An indication of this coupling in Tb is found in the tendency of $\mathcal{C}(\vec{q})$ and $\mathcal{D}(\vec{q})$ to oscillate strongly in a correlated fashion around A. The spin-orbit coupling mechanism, which gives rise to the violation of the selection rules for the magnon-phonon interaction, may introduce an interaction between the acoustic and optical magnons proportional to $\sin 3\phi$ instead of $\cos 3\phi$. This is the only modification introduced by this mechanism if the moments are parallel to an a or a b axis.

The order of magnitude of the two-ion anisotropy deduced in Tb, Dy, and Er suggests that its microscopic origin lies in the mechanisms which affect the indirect RKKY exchange interaction [mechanisms (i)–(iii)]. As we have stressed, the indications of contributions from the spin-orbit coupling of the conduction electrons, (ii), show that a nonrelativistic theory, (i), may only succeed in explaining part of the two-ion anisotropy observed in these metals.

The phenomenological spin-wave theory for a basal plane ferromagnet presented above accounts for the two-ion parameters which have been determined in Tb. According to the selection rules, Eqs. (26) and (27), $\mathcal{C}(\vec{q})$ may be connected with terms like $K_{044}^{033}(\vec{q})$ or $K_{035}^{024}(\vec{q})$ ($l+m$ and $l'+m'$ both odd, $\mu+m+m'=6$). The theory of Callen and Callen,[39] Eqs. (32) and (33), predicts a renormalization of these terms within the range $\sigma^{19}-\sigma^{35}$, which is in fair agreement with the experimental value of $k(\vec{q})$. $\mathcal{K}(\vec{q})$, the axial anisotropy term in $A_{\vec{q}}-B_{\vec{q}}$, may include contributions from $K_{02\,2}^{02\,-2}(\vec{q})$ ($\sim\sigma^5-\sigma^9$), which has been shown to be the most important coupling in the description of the spins in Er.[31]

The presence of two-ion anisotropy will in general affect the spin-wave energy gap at zero wave vector (III). An order of magnitude estimate may be based on the Fourier coefficients of the anisotropy parameters given in Table I. A comparison of Eqs. (22) and (26) shows that the interplanar contribution to $A(0)+B(0)$ of the two-ion couplings appearing as $\mathcal{C}(\vec{q})$ is determined by

$$-\sum_n \mathcal{C}_n(-\cos 6\phi) = -(1.6\ \text{meV})(-\cos 6\phi)$$

at zero temperature. Although the intraplanar contributions may be of the same order of magnitude, this number indicates a very strong correlation between $\mathcal{C}(\vec{q})$ and the surprisingly large anisotropy parameter, $\Delta M=-(1.39\ \text{meV})\sigma^{14.3}$, obtained in III. In III we discuss further evidence for the two-ion origin of ΔM. The connection between the two-ion contributions to $A(0)\pm B(0)$ and the other $\vec{q}$-dependent anisotropy parameter, $\mathcal{K}(\vec{q})$ and $\mathcal{D}(\vec{q})$, is not as close as in the case of $\mathcal{C}(\vec{q})$, because it depends on the types of coupling which are considered. If we assume that $m\approx m'$ for the terms which contribute to $\mathcal{D}(\vec{q})$ then the corresponding interplanar contribution to $A(0)-B(0)$ would be

$$-2\sum_n \mathcal{D}_n(-\cos 6\phi) = (0.2\ \text{meV})(-\cos 6\phi)\ .$$

If the axial anisotropy term in $A_{\vec{q}}-B_{\vec{q}}$ is dominated by a certain spin coupling then the interplanar contribution to the ϕ-independent part of $A(0)+B(0)$ would be negative [e.g., if $K_{02\,2}^{02\,-2}(\vec{q})$ dominates, the contribution is -1.6 meV]. In other cases the contribution to $\mathcal{K}(\vec{q})$ is quite uncertain.

Some of the arguments in this discussion are further elucidated in the following two articles, II and III. A summary of all three papers is given at the end of III.

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